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Publication date:
1966

Document Version
Publisher's PDF, also known as Version of record

Risö Report No. 141

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Danish Atomic Energy Commission

Research Establishment Risö

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September, 1966

Sales distributors: Jul. Gjellerup, 87, Sölvgade, Copenhagen K, Denmark

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Accelerator Section

Abstract

The suitability of a homopolymer type of PVC film (QCA-5960, 0.015" thick) as a high-dose electron dosimeter (1-5 Mrads) has been investigated. Different heat treatments were applied in order to obtain a reasonable storage stability and an approximately linear relationship between optical density and dose over the dose range from 0.5 to 5 Mrads. The spectral range was 3900 to 5000 Å. The effect of the dose rate on the coloration of PVC was studied.

CONTENTS

INTRODUCTION

Polyvinyl chloride (PVC) is coloured when exposed to ionizing radiation. The coloration increases with dose and is further developed and stabilized by means of a suitable heat treatment. PVC film may therefore serve as a dosimeter for gamma and electron rays and has been used for such purposes in our laboratory for some years. Work on a copolymer PVC (Union Carbide Bakelite Co.) has been published by J. E. Maul, N. W. Holm and J. G. Draganic in a Risö Report[1] and in a laboratory manual for high-level dosimeters[2]. These studies resulted in the recommendation of a heat treatment at 80°C for 30 minutes in order to obtain a coloration that is stable for two days.

The calibration curve, i. e. the relationship between the dose and the OD of the film, corresponding to this treatment is not linear. A linear relation between OD and dose would be desirable both in routine dosimetry and in experimental work.

We have extended our studies to a homopolymer PVC produced by Union Carbide Bakelite Co., which Artandi and Stonehill[3] have reported to render a linear OD-dose relationship.

The irradiation of PVC film is carried out in two different ways:

(1) Total-dose irradiation (the total dose is given in one irradiation. Used in routine dosimetry).

(2) Step-dose irradiation (the total dose is given by several irradiations with a constant dose (step dose)).

EXPERIMENTAL DETAILS

Radiation Source

The radiation source is an electron linear accelerator of the travelling wave type, operating at 3000 Mhz. The average power of the beam is 5 kW at an energy of 10 MeV. The electron energy is variable from 2 to 15 MeV. The pulse, which is of rectangular shape, has peak currents of 0.4 A at 5 MeV and 0.21 A at 10 MeV. The pulse duration can be varied from 0.2 to 7 μsec and the pulse repetition rate from 1.17 up to 300 pulses per second.

The beam can be directed through a straight-ahead window or through a 90° bending system. Below the bending system a scanning system with a scanner magnet is fitted to give a 40 cm spread of the beam in a direction perpendicular to that of the conveyor. For a given setting of the accelerator parameters, this allows a continuous range of doses from 15 krads to 45 Mrads per passage.

PVC Material

The PVC film studied is a rigid, non-plasticized homopolymer of polyvinyl chloride, produced by Union Carbide Plastics Division.

An examination of the thickness variation of the film (QCA 5960, clear 151 rigid vinyl sheets, 0.015 inches) gave the following results:

A. Thickness measurement across the sheet: Mean value 0.3489 mm, standard deviation 0.0095 mm.

B. Thickness measurement along the edge of the sheet: Mean value 0.3463 mm, standard deviation 0.0097 mm.

It is worth noting that the variation in thickness is most pronounced along the edges of the sheet in both directions.

Experimental Set-up

The PVC film was cut into strips, 8 mm by 280 mm. The strips were placed between two thin sheets of cardboard on top of the calorimeter used for dose determination. This set-up was placed in an aluminium tray and passed through the irradiation field by means of the conveyor.

Dose Measurement

A water calorimeter made of a thin-walled polystyrene Petri dish, 6" diameter and 9/16" thick, filled with water and embedded in polystyrene foam, was used. The temperature increase in the system was measured by means of a calibrated thermistor placed at the centre of the box. The reproducibility of the water calorimeter is better than 2%[5].

Measurement of Optical Density

The films were measured in a Zeiss PMQ II spectrophotometer specially supplied with a film-conveyor system allowing the strips to be recorded as they pass continuously through the monocromatic light beam. Before the measurement, the PVC films were cleaned carefully with lens paper. The colour intensity was measured against an unirradiated and unheated film at 3960 Å and 4800 Å two hours after heat treatment.

Irradiation Procedure and Heat Treatment

The coloration taking place during the irradiation is dependent upon the irradiation temperature[1]. It may therefore be advantageous to irradiate the PVC films by steps rather than give the total dose in one passage. The OD's of PVC films irradiated with the same dose by total-dose and step-dose irradiation respectively are different, the difference depending on the wave length used for spectrophotometric measurements. Consequently two groups of experimental results are obtained, related to the two different methods of irradiation.

In spite of the known dependence on the irradiation temperature no special care was taken to control the temperature during the irradiation, as it was preferred to calibrate under natural working conditions, starting at room temperature. For the heat treatment three different procedures were applied:

(1) PVC films were heated immediately after irradiation.

(2) All PVC films in one series of experiments were heated simultaneously after all irradiations were completed.

(3) All PVC films in one series of experiments were heated simultaneously three hours after the mean time between the first and the last irradiation in the series.

In the total-dose irradiations, PVC strips were given doses of 0.5, 1.0, 1.5, ... 5.0 Mrads.

In the step-dose irradiations, step doses of the order of 0.3 and 0.6 Mrad were used. Most emphasis was put on the 0.3 Mrad step dose, which was used for measurement of depth dose distribution in different materials. Low step doses are expedient in order to minimize heating of the sample. The best procedure is to give the first step dose to one PVC film and to add a film for each following irradiation. At the end of the irradiations the first PVC film has received the total dose desired, and the last film only 0.3 Mrad. This procedure provides an opportunity to heat all the PVC foils simultaneously after the entire irradiation, which is important from the point of view of standardized working conditions. In both procedures the film was heat treated in a temperature-controlled oven with forced air circulation. The temperature control was better than $\pm$ 2°C.

EXPERIMENTAL RESULTS

Absorption Spectra of PVC Film

Fig. 1 shows the spectrum of a homopolymer polyvinyl-chloride film irradiated with a dose of about 3 Mrads and heat treated at 60°C for 25 minutes. Absorption maxima appear at 3960, 4280, 4550, and 4800 Å.

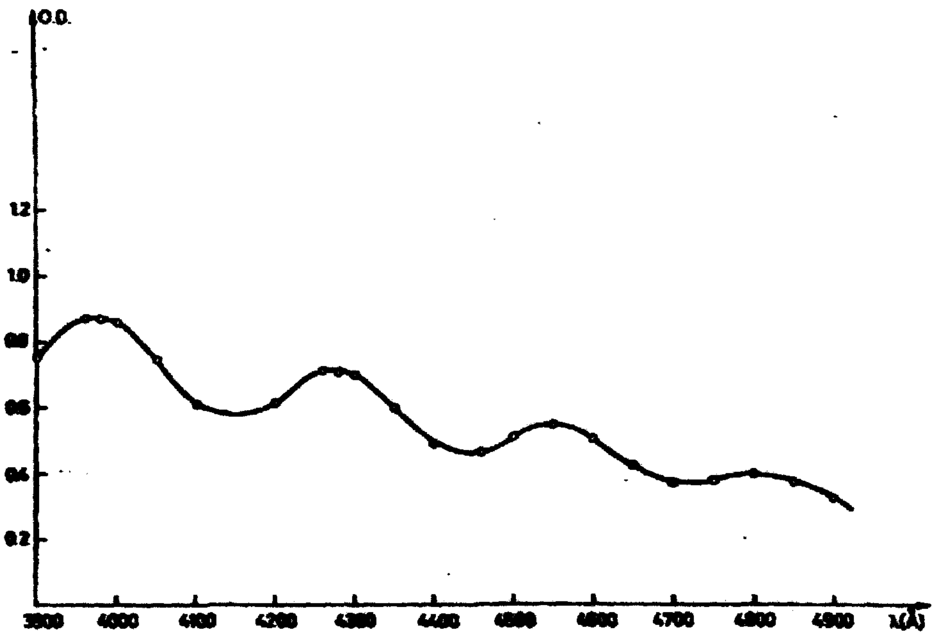

Fig. 1. Spectrum of PVC irradiated with dose 4 Mrads. Heat treatment applied immediately after irradiation: 15 min. at 60°C. OD was measured about 30 minutes after heating.

It is seen from the figure that several absorption peaks could be used in routine work. However, in checking the linear relationship between OD and absorbed dose, and the stability of the coloration after heat treatment, it was found that the peak at 4800 Å is to be preferred in spite of the comparatively low OD (see pages 10 and 11).

Heat Treatment

Heat treatments of different durations were given at the temperatures 50, 60, 70, and 80°C, and spectrophotometric measurements were made of the wave lengths 3960, 4280, 4550, and 4800 Å, in order to select the optimum fading characteristics combined with a reasonable coloration. The temperature 60°C gave the best response in this respect independently of the wave length, as seen in fig. 2.

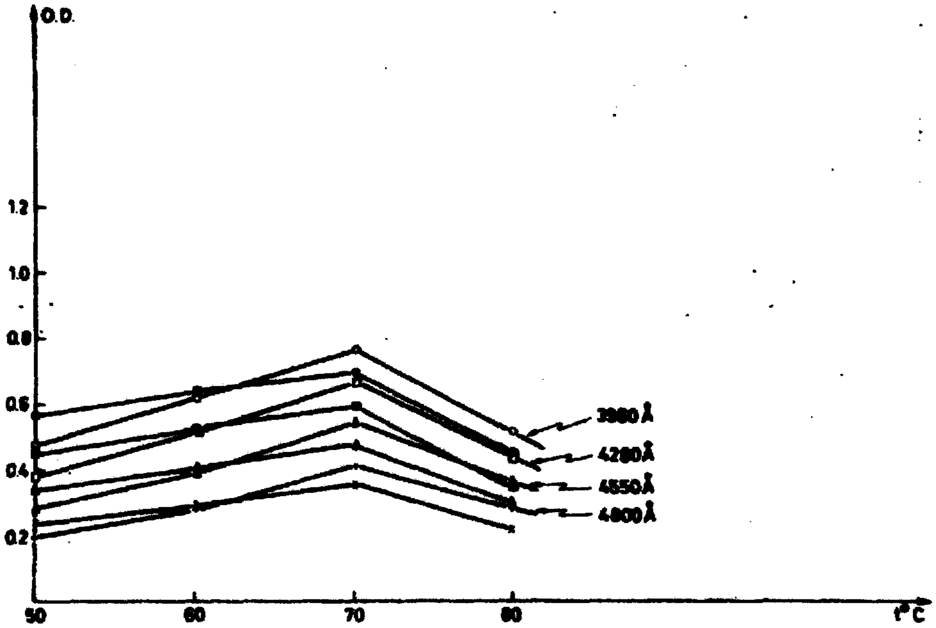

Fig. 2. Optical density of PVC at different wave-lengths in the temperature range from 50 to 80°C. 3960 Å - o, q; 4280 Å - ◻, ■; 4550 Å - △, ▲; 4800 Å - +, x. The first marks are for OD as measured 3 hours after heating, the second for measurement 20 hours after heating.

Different heating periods at 60°C were tried in order to obtain one that would keep the linearity between OD and absorbed dose and maintain the stability of OD within a reasonable time. Fig. 3 shows the calibration curves for applied heat treatments of 25, 35 and 60 minutes and the wave lengths 3960 and 4800 Å for the spectrophotometric measurements.

The time exposure of 25 minutes seemed best suited although the OD increased slightly within two hours after the heat treatment (see fig. 4).

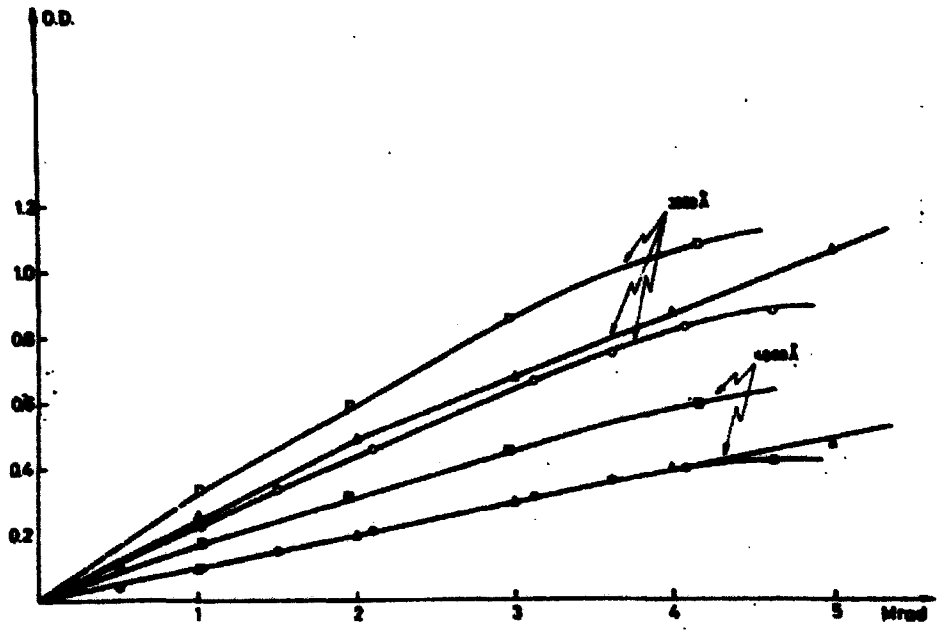

Fig. 3. Calibration curves for heat treatments of [illegible], [illegible], [illegible] minutes at wave-lengths [illegible] Å (Δ, o, ◻) and [illegible] Å (▲, •, ■).

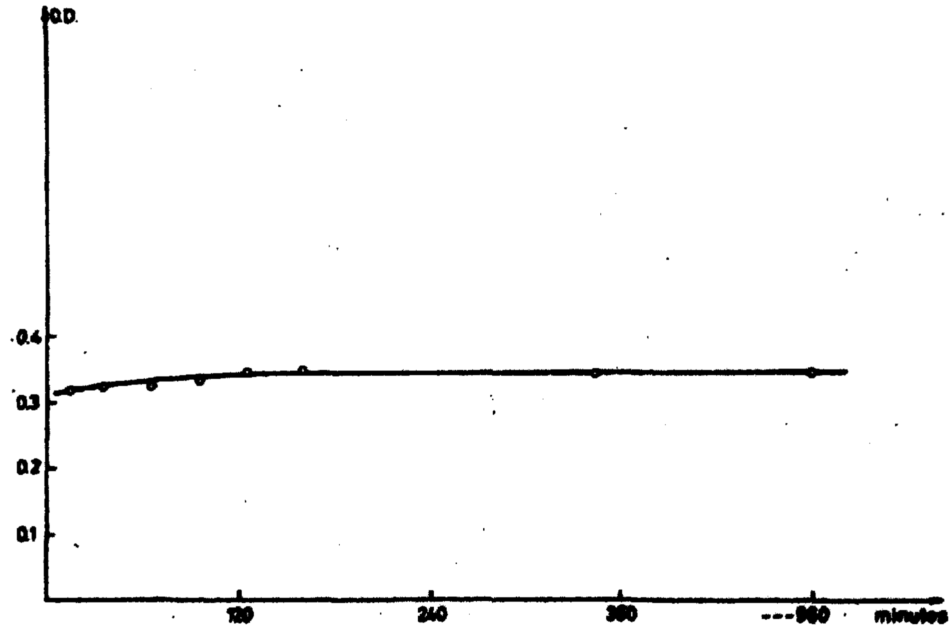

Fig. 4. Increase of OD within [illegible] hours after heat treatment for [illegible] minutes at [illegible]°C.

As, however, the increase of OD was completed within these two hours and OD did not vary for two days afterwards (see page 11), the time for spectrophotometric measurement can be fixed at two hours after completion of the heat treatment.

According to previous results[1, 6] we expected an influence of the dose rate on the OD of PVC films irradiated under atmospheric conditions. Changing the intensity of the electron beam by a factor of 10 from about 10^8 to about 10^9 rads/sec in the pulse, we examined the coloration of the PVC films. Standard conditions were used in a series of irradiations in order to make the data comparable. From figure 5 it is seen that a slight difference may exist, but it is insignificant for the given dose range and may properly be included in the experimental errors.

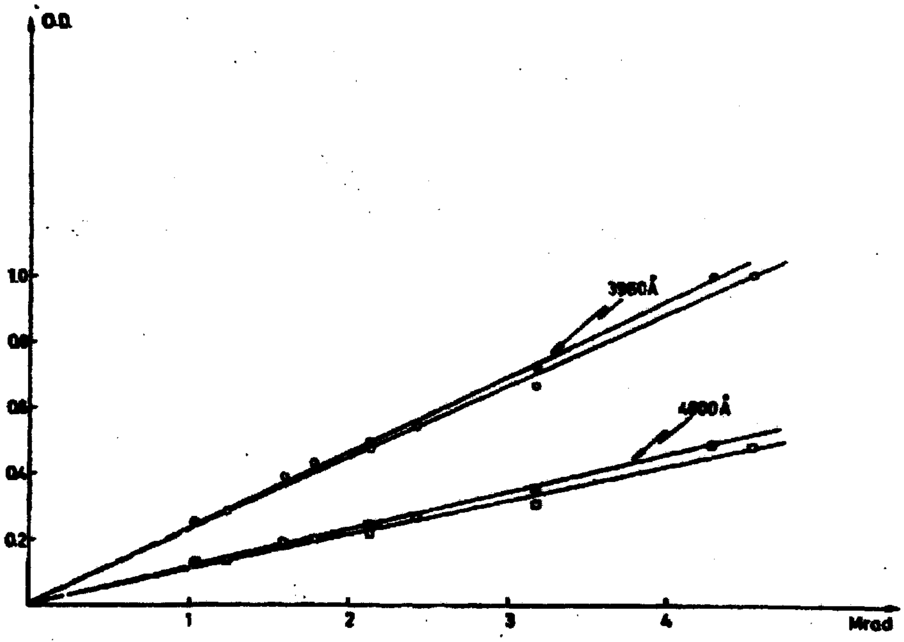

Fig. 5. Dose-rate influence on PVC. Low dose rate about 10^8 rads/sec. at wave-lengths 3960 Å - o and 4800 Å - □. High dose rate about 10^9 rads/sec. at wave-lengths 3960 Å - • and 4800 Å - ■.

Total-Dose Irradiation

As mentioned above, three different heat-treatment procedures were applied, and wave-lengths of 3960 and 4800 Å were used for the spectrophotometric measurement. We found that at the wave-length of 3960 Å, the OD and the linearity of OD versus absorbed dose depend on the time lapse between heat treatment and irradiation (see fig. 6).

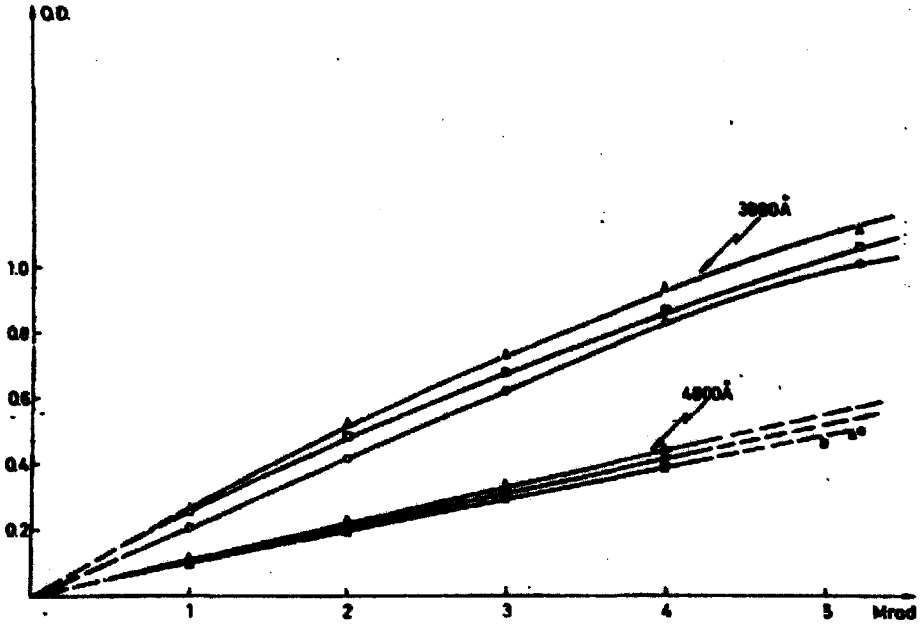

Fig. 6. Three different procedures for heat treatment after irradiation by total-dose method: 1. Heating the samples immediately after irradiation at wave-lengths 3960 Å - o, and 4800 Å - o. 2. Heating after entire irradiation with dose range up to 5 Mrads at wave-lengths 3960 Å - □ and 4800 Å - ■. 3. Heating 3 hours after irradiation at wave-lengths 3960 Å - ▲ and 4800 Å - ▲.

As seen in fig. 6, the heat treatment applied immediately after irradiation gives, at 3960 Å, a linear relationship up to 4 Mrads, while the two other treatments gives bent curves. These curves were obtained by heating all foils together after the entire experiment and with three hours' interval between irradiation and heat treatment.

At the wave length of 4800 Å the linear relationship between OD and absorbed dose was not disturbed within three hours by heating different periods of time after irradiation, although the slope is slightly changed (fig. 6). Stability of colour at 4800 Å was obtained within two days after heat treatment, while at 3960 Å some fading effect was seen on measurement 26 hours after heating (see fig. 7).

A straight-line relation was also obtained on irradiation in a Co^{60} facility (max. dose rate about 50 krads/min, dose range up to 4 Mrads, heat treatment 25 min. at $60^{\circ}C$ applied immediately after irradiation and at wave lengths of 3960 and 4800 Å), as can be seen in fig. 8. It is worth noting that the temperature of the irradiation facility was about $4^{\circ}C$.

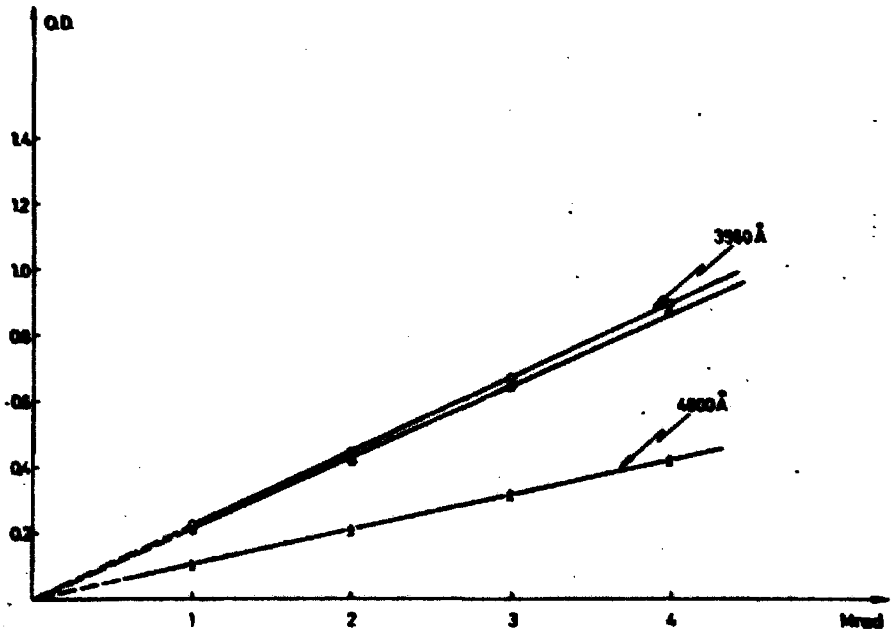

Fig. 7. Fading effect of total-dose irradiation. At wave-length 3960 Å, OD was measured 2 hours (o) and 20 hours (•) after irradiation, and at 4800 Å 2 hours (+) and 20 hours (x) after irradiation.

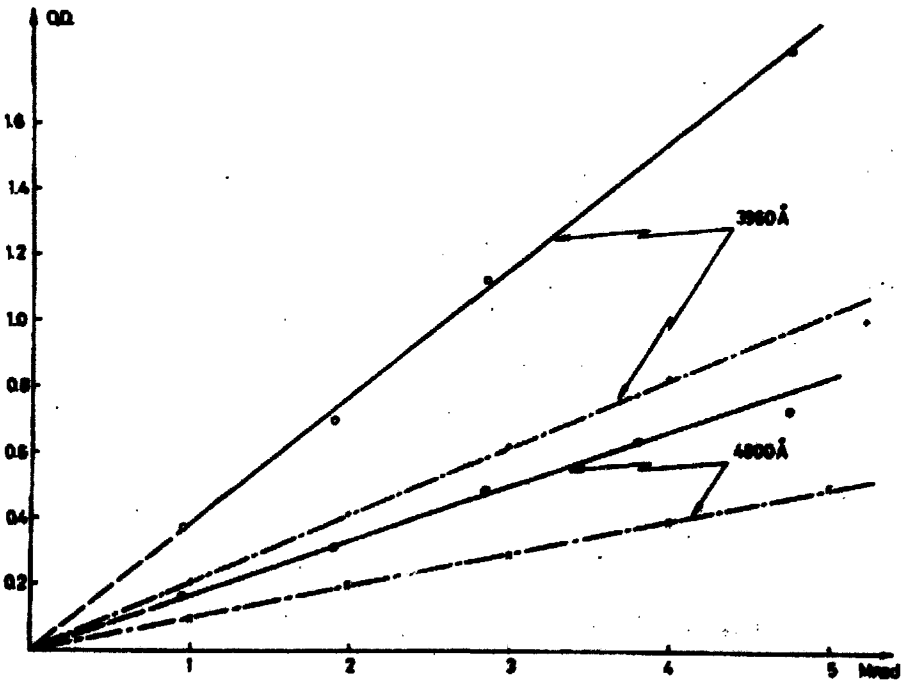

Fig. 8. Heat treatment for 25 minutes at 60°C applied to PVC films after irradiation by Co^{60} facility and electron beam. The curves are: at wave-length 3960 Å: o–Co^{60}, + – Acc; at 4800 Å: o–Co^{60}, x–Acc.

Step-Dose Method

When this method was used, the linearity of OD versus absorbed dose was shown to be independent of the three different heat-treatment procedures applied (a step dose of 0.3 Mrad was used, and measurements were taken at the wave-lengths 3960 and 4800 Å) (fig. 9). However, the slopes of the curves vary with the method of heat treatment at 3960 Å, but not so much at 4800 Å.

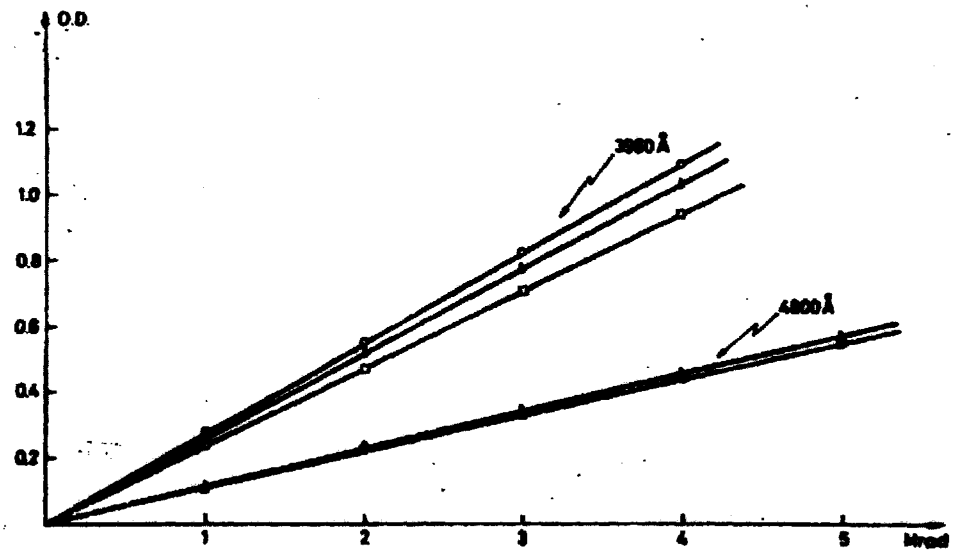

Fig. 9. Three different procedures for heat treatment after irradiation with step-dose of 0.3 Mrad: 1. Heating PVC films immediately after irradiation at wave-lengths 3960 Å - o and 4800 Å - o. 2. Heating after entire irradiation with dose range up to 5 Mrads at wave-lengths 3960 Å - □ and 4800 Å - ■. 3. Heating 3 hours after irradiation at wave-lengths 3960 Å - ▲ and 4800 Å - △.

With step doses of 0.6 Mrad and a wave-length of 3960 Å, some scattering occurred in the range between 2 and 4 Mrads, but still a straight line seems to apply (fig. 10).

It is seen from figure 11 that a better calibration curve is determined at 4800 Å, with a negligible fading effect for a step dose of 0.3 Mrad. No fading effect is observed at either wave length after 24 hours, but after 46 hours there may be a small effect, most pronounced at 3960 Å.

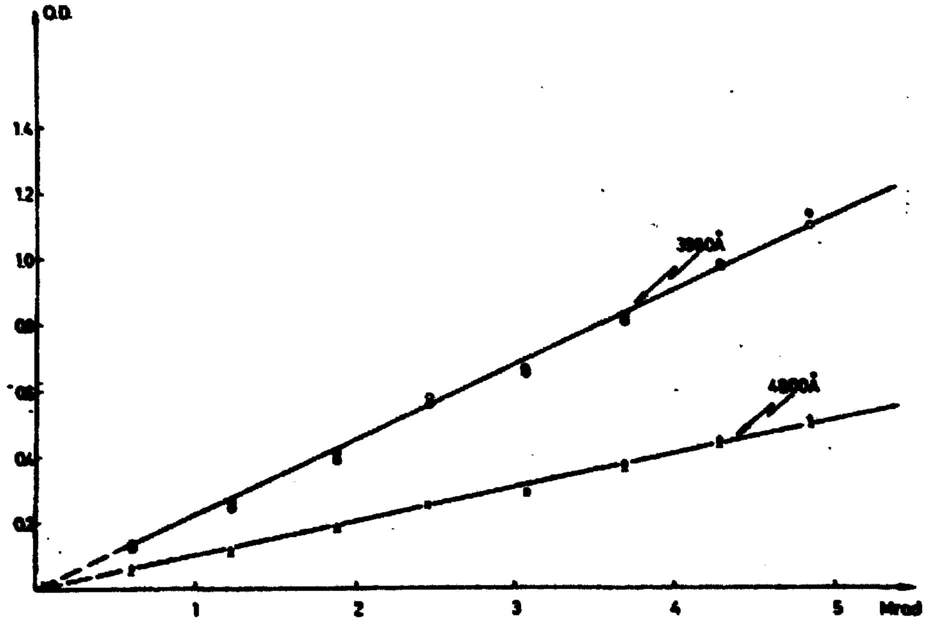

Fig. 10. Step-dose about 0.6 Mrad and heat treatment applied after entire irradiation. OD was measured 2 hours (o) and 22 hours (●) after heating at wave-length 3060 Å. Further, OD was measured at wave-lengths 4000 Å –+ (2 hours) and x (22 hours).

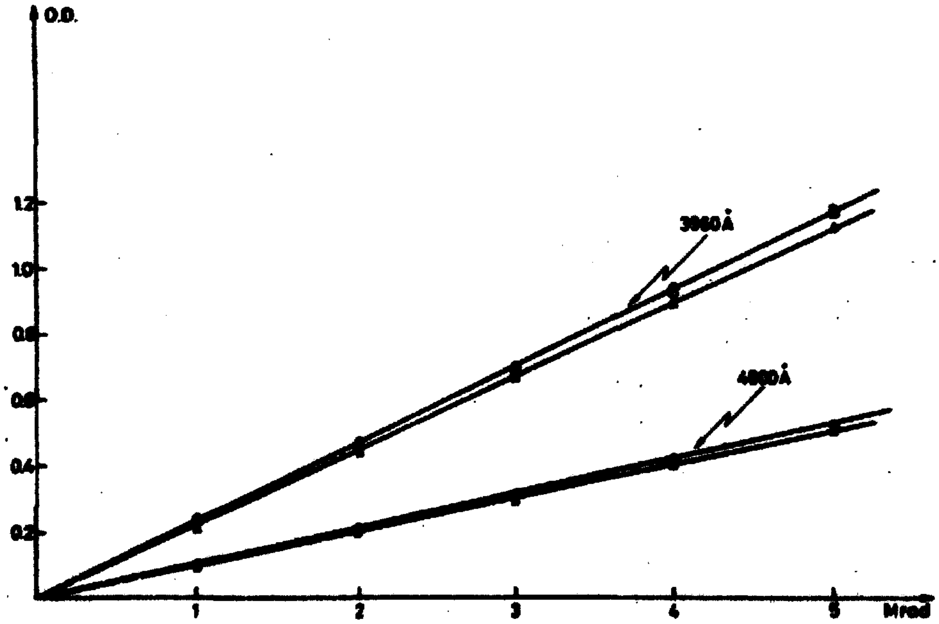

Fig. 11. Step-dose 0.3 Mrad and stability of OD within 26 hours at both wave-lengths. OD was measured after 2 hours (o), 26 hours (□) and 46 hours (Δ) at wave-length 3060 Å, and after 2 hours (o) and 46 hours (Δ) at wave-length 4000 Å.

CONCLUSION

It has been shown that the linear relationship between OD and absorbed dose, as well as the colour stability, can be sustained within a definite time by suitable heat treatment after irradiation. The following factors, however, influence the behaviour of PVC:

(1) Method of irradiation (total-dose or step-dose irradiation).

(2) Heat treatment (temperature (60°C) and time exposure (25 min)).

(3) Time when heat treatment is applied (see page 10).

(4) Wave-length used for the spectrophotometric measurement (3960 and 4800 Å).

(5) Time when OD is measured; may be more, but never less than two hours after heat treatment.

(6) Time expended on irradiation of one set of experiments - this time depends on which procedure of heat treatment is applied - (for the total-dose method, on an average about 50 minutes in the dose range from 0.5 to 4 Mrads; for the step-dose method maximum about 60 minutes in the dose range up to 5 Mrads when increments of 0.3 Mrad are used; with increments of 0.6 Mrad in the same dose range, about 50 minutes).

All these factors must be specified in order to maintain the linearity between the absorbed dose and the OD and to keep the stability of the intensity of the colour during a certain time.

Total-dose irradiation is suitable for routine work. The linear calibration curve without fading effect within 50 hours can be sustained after heat treatment by using a wave-length of 4800 Å for spectrophotometric measurement, and all the parameters mentioned. Caution should be exercised in reading the calibration curve as the OD is rather small and may easily be misread. The reproducibility is better than ± 6%, as can be seen from table I. A wave-length of 3960 Å is much better from the point of view of coloration, but in order to obtain a linear calibration curve the PVC film has to be heated immediately after irradiation. Further, the OD has to be measured within a much shorter time (not until two hours after heat treatment) because the fading effect appears 26 hours after heating.

In the case of the total-dose method, the usable dose range keeping linearity for both wave-lengths is limited to 4 Mrads.

The step-dose irradiation is not a convenient method for routine work since 0.3 Mrad is a very low step dose so that 16 consecutive irradiations have to be applied to give a total dose of 5 Mrads. For experimental work, however, it is very suitable, and it has been used in our laboratory for measurement of depth dose distribution in different materials. When this method is used, heating of the samples can be minimized[4]. The usable dose range is up to 5 Mrads, and the fading effect does not appear within 24 hours after heat treatment; even after 46 hours it is not higher than 5% for both wave-lengths used (see fig. 11). The wave-length of 4800 Å is preferable in this case also, since the slopes of the calibration curves are nearly the same for all three different treatments (see fig. 9).

The reproducibility is better than ± 5%, as can be seen from table II. The wave-length of 3960 Å can be used, but the time when heat treatment should be applied must be chosen within three hours after irradiation. This is necessary since the OD's of PVC films heated in the three different ways mentioned on page 5 show differences of about 16% within three hours after irradiation, as is seen in fig. 9.

ACKNOWLEDGEMENTS

The author is indebted to N. W. Holm and K. Sehested for many helpful discussions on the work and for checking of the manuscript. Further, thanks are due to the staff of the Accelerator Section, who made a great contribution to the performance of the experimental work.

REFERENCES

1) J.E. Maul, N.W. Holm and J.G. Draganic, The Use of Polyvinyl-Chloride Film for Co^{60} Radiation Dosimetry. Risö Report No. 31 (1961).

2) J.G. Draganic, N.W. Holm and J.E. Maul, Laboratory Manual for Some High-Level Chemical Dosimeters, Ferrous Sulphate, Oxalic Acid, Ceric Sulphate, Polyvinyl-Chloride Foils. Risö Report No. 22 (1961).

3) Charles Artandi and Albert A. Stonehill, Nuclear Instruments and Methods 6 (1960) 279-282.

4) Ari Brynjolfsson and Gunnar Thaarup, Determination of Beam Parameters and Measurements of Dose Distribution in Materials Irradiated by Electrons in the Range of 6 MeV to 14 MeV. Risö Report No. 53 (1963).

5) Ari Brynjolfsson, Niels W. Holm, Gunnar Thaarup, and Knud Sehested, Industrial Sterilization Processing at the Electron Linear Accelerator Facility at Risö. Conference on the Application of Large Radiation Sources in Industry, IAEA, Salzburg, 27-31 May, 1963.

6) A.A. Miller, Radiation Chemistry of Polyvinyl Chloride. J. Phys. Chem. 63 (1959) 1755.

$t_{irrad.}$ = time (minutes) spent on the irradiation.

$n_{s.d.}$ = number of irradiations with constant step dose.

Table I

Total-dose irradiation (0.5 - 4 Mrads)

Exp. no.	$t_{irrad.}$	OD 1 Mrad	OD 2 Mrads	OD 3 Mrads	OD 4 Mrads
1	45	0.100	0.199	0.298	0.396
2	36	0.108	0.212	0.317	0.420
3	43	0.095	0.187	0.277	0.367
4	51	0.092	0.187	0.282	0.374
5	57	0.103	0.204	0.308	0.402
Mean value	46 min	0.0996	0.1978	0.2964	0.3908
Standard deviation		0.0063 ~ ± 6.38%	0.0108 ~ ± 5.46%	0.0169 ~ ± 5.70%	0.0214 ~ ± 5.46% Average ± 5.75%

Table II

Step-dose method (0.3 - 5 Mrads)

Exp. no.	$t_{irrad.}$	$n_{s.d.}$	OD 1 Mrad	OD 2 Mrads	OD 3 Mrads	OD 4 Mrads	OD 5 Mrads
1	51	12	0.110	0.209	0.304	0.401	0.490^{x}
2	51	12	0.107	0.212	0.315	0.419	0.522^{x}
3	45	14	0.104	0.211	0.318	0.423	0.530^{x}
4	58	16	0.115	0.230	0.340	0.454	0.567
5	59	16	0.116	0.227	0.334	0.441	0.551
Mean value	53 min		0.1104	0.2178	0.3222	0.4276	0.5320
Standard deviation			0.0051 ~ ± 4.66%	0.0098 ~ ± 4.53%	0.0146 ~ ± 4.54%	0.0204 ~ ± 4.79%	0.0293 ~ ± 5.50% Average ± 4.80%

^{x} extrapolated values